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Measurement and correlation of the liquid viscosity of *trans*-1-chloro-3,3,3-trifluoropropene (R1233zd(E))

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ABSTRACT

In 2015, many nations have reached the agreement to limit greenhouse gas emissions in the Paris Agreement on climate change. New working fluids with low global warming potential (GWP) has thus caused extensive interest, which can protect the environment and meet engineering performance requirements. *trans*-1-Chloro-3,3,3-trifluoropropene (R1233zd(E)) is one of the new working fluids that has potential applications in refrigeration systems. In this work, viscosity data of *trans*-1-chloro-3,3,3-trifluoropropene were reported. The viscosity was meausred over the temperature range of (243–373) K and at pressures up to 40 MPa using a vibrating-wire viscometer. The combined expanded uncertainty of the reported viscosity is 2% with a confidence level of 0.95 ($k = 2$). These experimental data were used to correlate a viscosity equation that covers a wide temperature and pressure range, with an estimated uncertainty at a 95% confidence level of 2% for the liquid phase from (240 to 400) K at pressures up to 40 MPa.

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1. Introduction

In 1974, Molina and Rowland [1] found that chlorofluorocarbons (CFCs) and various other halogenated compounds were destroying the Earth's stratospheric ozone. So, the replacements of CFCs have been developed and studied, such as hydrochlorofluorocarbons (HCFCs) and hydrofluorocarbons (HFCs). Unfortunately, the widely use of HCFCs and HFCs led to another environmental trouble of global warming. In order to deal with emissions of greenhouse gas, the Paris Agreement on climate change was reached within the United Nations Framework Convention on Climate Change (UNFCCC) in the 21th climate change conference in 2015, which indicated that many of the existing compounds in the refrigeration field with ozone depletion concerns and/or contributions to global warming are planned to be forbidden. New working fluids with low global warming potential (GWP) has thus caused extensive interest, which can protect the environment and meet engineering performance requirements. *trans*-1-Chloro-3,3,3-trifluoropropene (R1233zd(E)) is one of the new working fluids, which is nonflammable and low toxicity [2–4].

The destruction process of the CFC is photolysis by solar ultraviolet radiation in the stratosphere with essentially no removal process in the troposphere [5]. The main removal process for CFC substitutes in the troposphere is the reaction with hydroxyl radicals, which reduces their delivery to the stratosphere. The majority products following the OH radicals attack undergo fast chemical reactions or hydrolysis and dissolution in atmospheric water followed by their washing out from the atmosphere [5]. Sulbaek Andersen et al. [6] studied the kinetics of the gas-phase reactions of Cl, OH radicals and O_3 with R1233zd(E). Based on their results, Patten and Wuebbles [7] calculated the ozone depletion potential (ODP) of R1233zd(E) using a 3-D chemical-transport model of the atmosphere, and the value of ODP was 0.00034 with the atmospheric lifetime of 40.4 days. Recently, Orkin et al. [5] reported new OH reaction rate constant with R1233zd(E). The authors also estimated the atmospheric lifetime and the ODP, which are 46 days and 0.00050, respectively. Although R1233zd(E) contains one chlorine atom in the molecule, the contribution to stratospheric ozone depletion is expected to be negligible because of its very short-atmospheric lifetime. The global warming potential of R1233zd (E) is 7 relative to carbon dioxide at an integration time horizon of 100 years (GWP_{100}) [6].

It is essential to know the accurate thermophysical properties of the working fluids to optimize the design of energy-conversion systems and to select the alternative environmentally friendly working fluids. Some of the thermodynamic properties of R1233zd(E) had already been reported, including vapor pressure, density, speed of sound, ideal gas heat capacity, surface tension and thermal conductivity [8–16]. With regard to viscosity, to the

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best of our knowledge, only two data sets were reported in literature [8,16], with limited temperature and pressure ranges. For a more comprehensive knowledge of the fluid, in this work, the compressed liquid viscosities of R1233zd(E) were reported, measured by using a vibrating wire viscometer and covering a temperature range from (243 to 373) K and at pressures up to 40 MPa. Based on these accurate experimental data, a viscosity correlation of R1233zd(E) was developed.

2. Experimental section

2.1. Chemicals

trans-1-Chloro-3,3,3-trifluoropropene was provided by Honeywell International Incorporation, with a stated mass purity of 0.999, and was used without purification. We used a gas chromatography equipped with a thermal conductivity detector and a capillary column (60 m × 0.32 mm) to test the purity of the samples, with a result of GC area fraction of 0.999, which could be used to infer an approximate chemical purity. Table 1 lists the specification of the sample used in this work.

2.2. Apparatus

The vibrating wire viscometer in a steady-state mode used here has been described previously [17,18]. The viscometer consisted of a custom-made stainless steel vessel with a maximum design pressure of 70 MPa, a pair of 40 mm-length samarium-cobalt magnets and some other components. A tungsten wire clamped at both ends while maintaining the tension of the wire was used, with a nominal radius of (50.00 ± 0.32) μm and length of 58 mm. A sinusoidal voltage was applied on the wire immersed in the testing fluid provided by a function generator. The Lorentz force drove the wire's transverse oscillation motion, subject to a permanent magnetic field. The viscosity and density of the fluid affected the wire's oscillation, according to the Navier-Stokes equations. The electromotive force at the wire was affected by the oscillation motion, conversely. The faint in-phase and quadrature voltages across the wire were detected by a lock-in amplifier over the frequency range to calculate the viscosity. The measured complex voltages were recorded to fit the working equations, which included the parameters such as the known density and the unknown viscosity for each measuring point. The viscosity then can be calculated from the working equations. Some of the system's parameters were obtained from the calibration and validation, which were discussed previously [18] without change. Before the measurement, the system was evacuated to avoid the impurities (such as air) in the cell. The wasting vessel was used to collect the fluid and the system was washed by solvents such as ethanol and acetone after the measurement.

A thermostatic bath provided the constant temperature of the viscometer and a calibrated 100 Ω platinum resistance thermometer was used to measure the temperature, connected to a digital multimeter. The combined expanded uncertainty of temperature was $U_c(T)$ = 12 mK, with a level of confidence of 0.95 (k = 2). The pressure was generated by a manual piston pump, and was measured by a high-pressure transducer with a pressure range up to 100 MPa. A nanovolt meter with 7 1/2 digits resolution was used to measure the pressure transducer signal. The combined expanded uncertainty of pressure $U_c(p)$ = 0.12 MPa (k = 2). Considering the uncertainties of temperature, pressure, repeatability of measurement, and the density of the fluid calculated from the equation of state [10], the combined expanded uncertainty of viscosity with a level of confidence of 0.95 (k = 2) was better than 2%.

Table 1
Specification of samples.

Chemical name	CAS No.	Formula	Initial mass fraction purity	GC area fraction
R1233zd(E)	102687–65-0	$C_3ClH_2F_3$	0.999	0.999

3. Experimental results

The compressed liquid densities for R1233zd(E) between (243 to 373) K and at pressures up to 40 MPa were calculated from the correlation proposed by Mondéjar et al. [10], with the uncertainty of 0.02%. The viscosities of compressed liquid R1233zd(E) were measured at pressures from (0.2 to 40) MPa, along 14 isotherms from (243 to 373) K, listed in Table 2. The viscosity data as a function of pressure at different temperatures are shown in Fig. 1.

Table 2
Viscosity η of R1233zd(E) at temperature range of (243 to 373) K from (0.2 to 40) MPa measured using a vibrating wire viscometer. [a]

T/K	p/MPa	ρ[b]/kg·m^{-3}	η/mPa·s
243.15	0.2	1387.7	0.5325
243.15	1.0	1389.1	0.5420
243.14	5.0	1395.7	0.5536
243.15	10.0	1403.7	0.5859
243.15	20.0	1418.4	0.6281
243.15	30.0	1431.8	0.6899
243.15	40.0	1444.2	0.7324
253.15	0.2	1366.1	0.4733
253.15	1.0	1367.6	0.4782
253.15	5.0	1374.9	0.4887
253.15	10.0	1383.5	0.5179
253.14	20.0	1399.4	0.5583
253.15	30.0	1413.7	0.6008
253.15	40.0	1426.9	0.6521
263.14	0.2	1344.1	0.4168
263.15	1.0	1345.7	0.4194
263.15	5.0	1353.7	0.4330
263.15	10.0	1363.1	0.4550
263.15	20.0	1380.2	0.4998
263.15	30.0	1395.6	0.5390
263.16	40.0	1409.6	0.5767
273.15	0.2	1321.6	0.3733
273.15	1.0	1323.5	0.3741
273.15	5.0	1332.3	0.3892
273.15	10.0	1342.5	0.4091
273.15	20.0	1361.0	0.4448
273.14	30.0	1377.5	0.4792
273.15	40.0	1392.4	0.5197
283.14	0.2	1298.7	0.3347
283.15	1.0	1300.7	0.3363
283.15	5.0	1310.4	0.3510
283.14	10.0	1321.6	0.3688
283.15	20.0	1341.7	0.4033
283.15	30.0	1359.4	0.4368
283.15	40.0	1375.2	0.4708
293.15	0.2	1275.1	0.3004
293.15	1.0	1277.3	0.3033
293.15	5.0	1288.1	0.3160
293.15	10.0	1300.4	0.3332
293.14	20.0	1322.2	0.3656
293.15	30.0	1341.2	0.3971
293.14	40.0	1358.0	0.4280
303.15	0.2	1250.7	0.2719
303.15	1.0	1253.3	0.2743
303.15	5.0	1265.3	0.2874
303.15	10.0	1278.9	0.3033
303.15	20.0	1302.6	0.3337
303.15	30.0	1322.9	0.3635
303.16	40.0	1340.9	0.3930
313.15	0.3	1225.9	0.2464
313.16	1.0	1228.4	0.2489

(*continued on next page*)

Table 2 (*continued*)

T/K	p/MPa	ρ[b]/kg·m^{-3}	η/mPa·s
313.16	5.0	1241.9	0.2611
313.15	10.0	1256.9	0.2767
313.15	20.0	1282.7	0.3058
313.15	30.0	1304.6	0.3337
313.15	40.0	1323.7	0.3611
323.15	0.4	1200.2	0.2243
323.15	1.0	1202.6	0.2267
323.15	5.0	1217.8	0.2393
323.15	10.0	1234.5	0.2540
323.15	20.0	1262.7	0.2819
323.15	30.0	1286.2	0.3084
323.14	40.0	1306.5	0.3345
333.15	0.5	1173.3	0.2047
333.15	1.0	1175.7	0.2069
333.15	5.0	1193.0	0.2192
333.15	10.0	1211.6	0.2344
333.15	20.0	1242.4	0.2620
333.15	30.0	1267.7	0.2888
333.15	40.0	1289.3	0.3147
343.15	0.6	1145.1	0.1887
343.15	1.0	1147.3	0.1903
343.15	5.0	1167.2	0.2029
343.15	10.0	1188.1	0.2179
343.15	20.0	1221.9	0.2450
343.14	30.0	1249.1	0.2702
343.14	40.0	1272.1	0.2946
353.15	5.0	1140.3	0.1846
353.14	10.0	1163.9	0.1999
353.15	20.0	1201.1	0.2277
353.14	30.0	1230.4	0.2503
353.15	40.0	1254.9	0.2733
363.15	5.01	1112.1	0.1701
363.15	10.0	1139.0	0.1845
363.15	20.0	1180.0	0.2107
363.16	30.0	1211.6	0.2345
363.15	40.0	1237.7	0.2570
373.15	5.00	1082.4	0.1581
373.15	10.0	1113.3	0.1734
373.15	20.0	1158.6	0.1996
373.15	30.0	1192.7	0.2229
373.14	40.0	1220.4	0.2449

[a] The combined expanded uncertainty $U_c(k=2)$ are $U_c(T)$ = 12 mK, $U_c(p)$ = 0.12 MPa and $U_c(\eta)$ = 2% with confidence level of 0.95.
[b] Density ρ listed above was calculated from Mondéjar et al. [10].

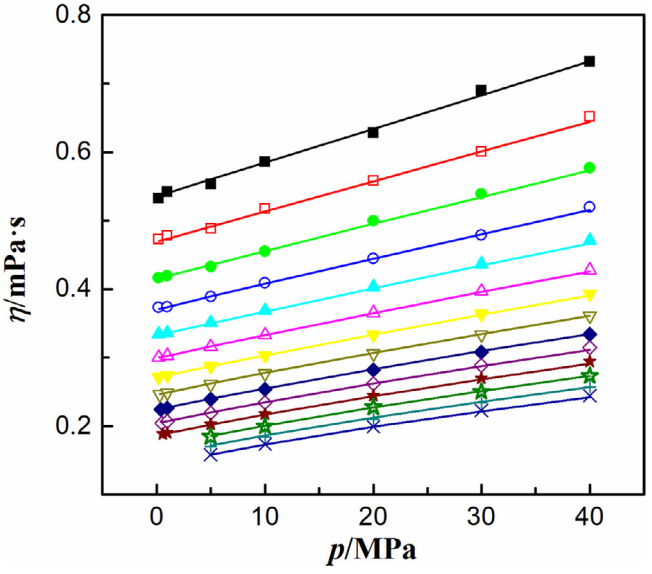

Fig. 1. Viscosity of R1233zd(E), η, as a function of pressure at different isotherms. (■) 243 K, (□) 253 K, (●) 263 K, (○) 273 K, (▲) 283 K, (△) 293 K, (▼) 303 K, (▽) 313 K, (◆) 323 K, () 333 K, (★) 343 K, (☆) 353 K, (+) 363 K, (×) 373 K.

4. Viscosity correlation

The viscosity η can be expressed as a sum of four contributions [19]:

$$\eta(\rho,T) = \eta_0(T) + \eta_1(T)\rho + \Delta\eta(\rho,T) + \Delta\eta_c(\rho,T) \quad (1)$$

where T is the temperature in K, ρ is the density in kg·m^{-3}. The first term $\eta_0(T)$ is the dilute-gas limit viscosity, and the second term $\eta_1(T)$ is the first-density coefficient. $\eta_0(T)$ and $\eta_1(T)$ were obtained theoretically in this work, which would be discussed later. $\Delta\eta_c$ is the enhancement of the viscosity in the critical region, which is considered only at the region near the critical point and is ignored in this work because we have no experimental viscosity data existence near the critical point. $\Delta\eta$ is the residual viscosity, which can only be obtained by empirically fitting from the experimental data. The correlation proposed by Mondéjar et al. [10] was used to calculate the density, ρ for each experimental temperature and pressure point for R1233zd(E), which was available in the NIST REFPROP database program [20]. The critical parameters for R1233zd(E) are T_c = 439.6 K, P_c = 3.6237 MPa, ρ_c = 480.219 kg·m^{-3}, and the molar mass is 130.4944 g·mol^{-1} [10]. These parameters are listed in Table 3. The range of state points covered by the present measurements is shown in Fig. 2 relative to the vapor-pressure curve of R1233zd(E).

4.1. The dilute-gas limit and the initial-density dependence terms

The dilute-gas limit viscosity, $\eta_0(T)$, is only determined by the temperature, which can be expressed in engineering units as [21]

$$\eta_0(T) = \frac{0.02669 \times (M_w T)^{1/2}}{\sigma^2 \Omega^*} F_c \quad (2)$$

where η_0 is in μPa·s, M_w is the molar mass in g·mol^{-1}. σ is the Lennard-Jones collision diameter in nm

$$\sigma = 0.809 V_c^{1/3} \quad (3)$$

where V_c is the critical volume in cm^3·mol^{-1}. Ω^* is the dimenssionless Lennard-Jones collision integral, which can be obtained from the expirical correlation presented by Neufeld et al. [22]

Table 3
Coefficients and parameters for the viscosity correlation of R1233zd(E) for Eqs. (1)–(12).

The critical parameters	
T_c (K)	439.6
P_c (MPa)	3.6237
ρ_c (mol·L^{-1})	3.68
V_c (cm^3·mol^{-1})	271.739
The coefficients for Eqs. (2)–(7)	
M_w (g·mol^{-1})	130.5
σ (nm)	0.524
ε/k_B (K)	349.08
ω	0.3025
κ	0
μ (Debye)	1.44
The coefficients b_i for Eq. (10)	
b_0 = −19.572881	b_1 = 219.73999
b_2 = −1015.3226	b_3 = 2471.0125
b_4 = −3375.1717	b_5 = 2491.6597
b_6 = −787.26086	b_7 = 14.085455
b_8 = −0.34664158	
The coefficients c_i for Eq. (12)	
c_0 = 17.5983	c_1 = −4.02103
c_2 = -105.820	c_3 = −10.6936
c_4 = 6.57574	c_5 = −5.26734
c_6 = 1.10633	c_7 = −0.959157

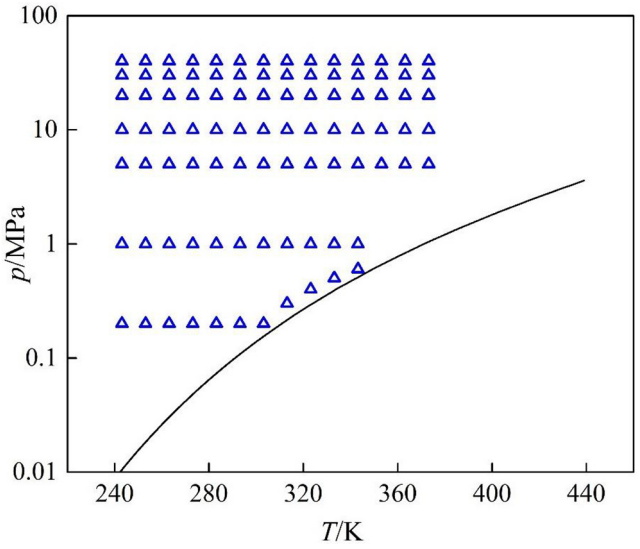

Fig. 2. Distribution of the data for the viscosity of R1233zd(E). △, experimental points measured in this work. The solid line shows the vapor–liquid saturation boundary.

$$\Omega^*(T^*) = 1.16145(T^*)^{-0.14874} + 0.52487e^{-0.7732T^*} + 2.16178e^{-2.43787T^*} - 6.435\times10^{-4}(T^*)^{0.14874} \times \sin[18.0323(T^*)^{-0.7683} - 7.27371] \quad (4)$$

where $T* = k_BT/\varepsilon$, ε is the Lennard-Jones energy parameter, and k_B is the Boltzmann constant. ε/k_B is expressed as

$$\varepsilon/k_B = T_c/1.2593 \quad (5)$$

In Eq. (2), F_c is used to correct the effect of non-smooth sphere molecular, which is given by

$$F_c = 1 - 0.2756\omega + 0.059035\mu_r^4 + \kappa \quad (6)$$

where ω is the acentric factor, κ is the correction factor for hydrogen-bonding effect of associating substances. κ equals to zero in this case. μ_r is a dimensionless dipole moment

$$\mu_r = 131.3\mu/(V_cT_c)^{1/2} \quad (7)$$

where μ is the dipole moment, T_c is the critical temperature in K. μ was estimated from B3LYP/aug-cc-pVTZ calculations using Gaussian 09 [23]. ω was given in the NIST REFPROP database program [20], where it was calculated from its definition using the equation of state of Mondéjar et al. [10]. With all the parameters known, Eqs. (2)–(7) can be used to calculate $\eta_0(T)$.

In order to show the temperature dependence of the term $\eta_1(T)$ in Eq. (1), we chose the model presented by Vogel et al. [24]. The second viscosity virial coefficient $B_\eta(T)$ was expressed as

$$B_\eta(T) = \frac{\eta_1(T)M_w}{\eta_0(T)} \quad (8)$$

The second viscosity virial coefficient can be calculated from the theory of Rainwater and Friend [25,26] as a function of a reduced second viscosity virial coefficient, $B_\eta^*(T^*)$, as:

$$B_\eta^*(T^*) = \frac{B_\eta(T)}{N_A\sigma^3} \quad (9)$$

where [27]

$$B_\eta^*(T^*) = \sum_{i=0}^{6} b_i(T^*)^{-0.25i} + b_7(T^*)^{-2.5} + b_8(T^*)^{-5.5} \quad (10)$$

In Eq. (9), N_A is the Avogadro's constant [28], 6.022140857 × 10^{23} mol^{-1}, while σ is the Lennard-Jones collision diameter in m, and M_w is the molar mass in kg·mol^{-1}. The coefficients b_i from Vogel et al. [27] are also shown in Table 3. It should be noted that we were unable to find any viscosity data in the gas phase, so the model is totally predictive for the gas phase and it is difficult to assess an uncertainty estimation; however the author's of this method [21] claim that comparisons with gas-phase data at low-pressure typically had an average absolute deviation of 1.5%. The method did not include fluorinated ketone compounds such as R1233zd(E) in its development, and the uncertainty in the gas phase is estimated to be about 10%.

4.2. The residual term

The residual viscosity term $\Delta\eta(\rho,T)$ is totally correlated from experimental data, because of the lack of a theoretical model. A symbolic regression software [29] was used to fit the experimental data in this work to determine the function and coefficients of the residual viscosity. Assael et al. [30] had suggested a form according to the hard-sphere model which was used in this work:

$$\Delta\eta(\rho_r, T_r) = (\rho_r^{2/3}T_r^{1/2})F(\rho_r, T_r) \quad (11)$$

where the functional form for $F(\rho_r,T_r)$ was entirely determined by the symbolic regression. For each experimental point, the dilute-gas limit $\eta_0(T)$ and the initial dependence terms $\eta_1(T)\rho$ were obtained from Eqs. (2)–(10). The residual term can be calculated by subtracting the $\eta_0(T)$ and $\eta_1(T)\rho$ from the experimental viscosity. A simple expression was found:

$$\Delta\eta(\rho_r, T_r) = \rho_r^{2/3}T_r^{1/2}\left(c_0 + \frac{c_1 + c_2\rho_r}{c_3 + c_4\rho_r + c_5T_r + c_6\rho_rT_r + c_7\rho_r^2}\right) \quad (12)$$

Coefficients c_i were listed in Table 3 for R1233zd(E). T_r is the reduced temperature $T_r = T/T_c$ and ρ_r is the reduced densty $\rho_r = \rho/\rho_c$, with $\Delta\eta$ in μpa·s.

4.3. Comparisons with experimental data

The viscosity data of 92 experimental points in the compressed liquid regions, from T = (243–373) K at pressures to 40 MPa, were used to develop a correlation for the viscosity of R1233zd(E). Fig. 3 shows deviations between the data and the correlation as a function of temperature. The average absolute percentage deviation, AAD, the maximum percentage deviation, MD, the average percentage deviation, Bias, and the relative standard deviation, RSD, were used to evaluate the correlation. η_{exp} is the experimental value of the viscosity, η_{cal} is the value calculated from the correlation, and the summation is over all n points. The AAD of the correlation is 0.4%, and the Bias is 0.05%. To the best of our knowledge, the viscosity data for R1233zd(E) in the public literature was from Hulse et al. [8] and Cui et al. [16]. Fig. 4 shows the deviations, as a function of density, of the experimental data of this work, Hulse et al. [8] and Cui et al. [16] from our correlation (Eqs. (1)–(12)). Cui et al. [16] used a surface light scattering method which was the same experimental system as described previously [31], at temperatures of 303 to 373 K. The kinematic viscosity uncertainty is about 2% with a confidence level of 0.95 (k = 2) according to the author's judgment. The maximum deviation of the values from Cui et al. [16] is 4.67%, which we considered to be good agreement. Hulse et al. [8] used a specially designed capillary tube method, and the deviation of the values from Hulse et al. [8] is from 22% to 45%. The data of the surface tension were also compared between Hulse et al. [8] and Cui et al. [16], with the deviation of about 10% to 20%. So, we considered that the data of Hulse et al. [8] may consist systematic deviation.

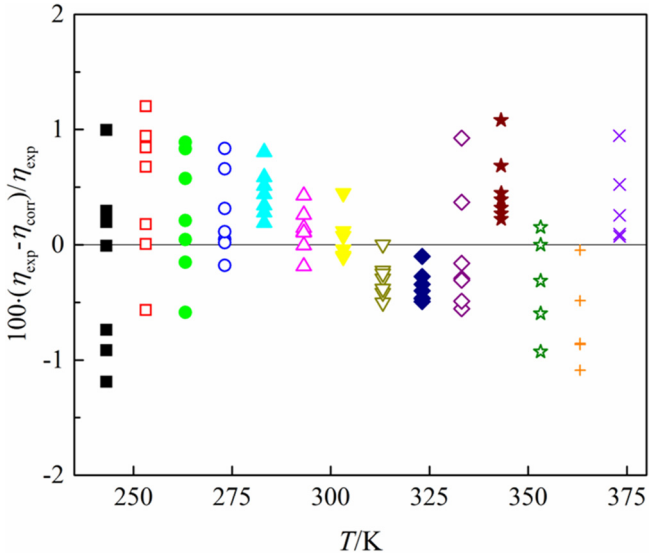

Fig. 3. Relative deviation between the present experimental data and the correlation for the viscosity of R1233zd(E) as a function of temperature for the liquid phase at pressures up to 40 MPa. (■) 243 K, (□) 253 K, (●) 263 K, (○) 273 K, (▲) 283 K, (△) 293 K, (▼) 303 K, (▽) 313 K, (◆) 323 K, () 333 K, (★) 343 K, (☆) 353 K, (+) 363 K, (×) 373 K.

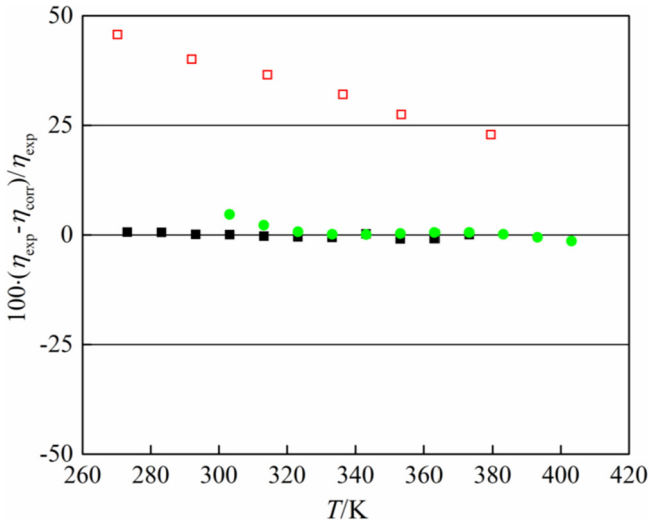

Fig. 4. Relative deviation between the present experimental data and the correlation for the viscosity of R1233zd(E) as a function of density for the liquid phase at pressures up to 40 MPa. (■) This work, (□)Hulse et al. [8], (●) Cui et al. [16].

The uncertainty of the correlation in the liquid phase over the temperature range 240 K–400 K at pressures up to 40 MPa is estimated to be as the same as that of the experimental data, namely 2%. The correlation may be extended to the part range of the equation of state of Mondéjar et al. [10], from (195 to 550) K at pressures up to 100 MPa, probably with the uncertainty of greater than 2%. Fig. 5 shows the smooth behavior of the correlation when extrapolated is demonstrated, which shows the viscosity of R1233zd(E) as a function of temperature at different pressures, with the temperature range from (200 to 1000) K and at pressures up to 50 MPa. Values for computer-program verification of the viscosity equation are given in Table 4, giving viscosity results calculated from the equation at different temperatures and densities.

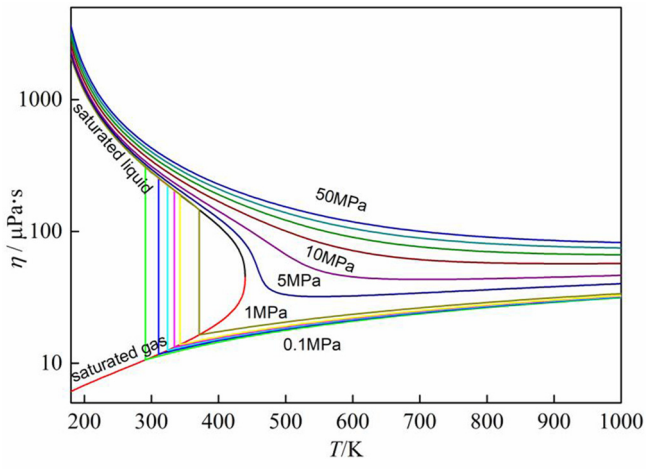

Fig. 5. Viscosity of R1233zd(E) as a function of temperature at different pressures.

Table 4
Sample points for computer verification of the correlating equation.

T/K	ρ/kg·m^{-3}	η/μPa·s
250	0	8.52
250	0.41	8.63
250	1809.77	4606.7
300	0	10.29
300	3.89	10.81
300	1701.48	2059.1
350	0	12.04
350	4.42	12.7
350	1595.99	1041.9

5. Conclusion

New viscosity data for *trans*-1-chloro-3,3,3-trifluoropropene (R1233zd(E)) have been presented in this work. A vibrating-wire viscometer was used to measure the viscosity at pressures up to 40 MPa and temperatures from (243 to 373) K. The combined expanded uncertainty of the results with a level of confidence of 0.95 (k = 2) is estimated to be about 2%. A viscosity correlation of R1233zd(E) was developed from these experimental data. The correlation may be used over the range of validity of the equation of state of Mondéjar et al., from (195 to 550) K at pressures up to 100 MPa. The estimated uncertainty of the correlation at a 95% confidence level is 2% for the liquid phase over the temperature range (240–400) K at pressures to 40 MPa.

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